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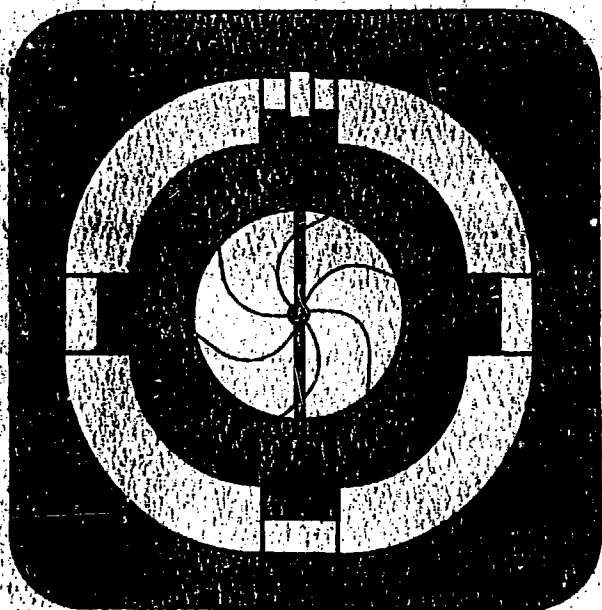
UNIVERSITY OF CALIFORNIA

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## MUON CATALYZED FUSION

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Contribution to Workshop on Fundamental Muon Physics  
Los Alamos, New Mexico, January 20-22, 1986

Presented by: W. H. Breunlich and P. Kammel

MUON CATALYZED FUSION

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I. INTRODUCTION

In recent years muon catalyzed fusion (MCF) has gained great interest due to the observation of resonance effects in formation of  $d_2d$  molecules<sup>1,2</sup> and the predictions of extremely fast rates in the deuterium-tritium (DT) cycle<sup>3,4</sup>. Even concepts to use MCF for energy production are being considered<sup>5</sup>. Currently several experimental programs are under way to study muon catalyzed DT fusion and first results indeed confirm the existence of a very rapid DT cycle<sup>6-9</sup> producing multiple fusions of the type



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This paper presents an overview of the program and results of our experiment performed by a European-American collaboration at the Swiss Institute of Nuclear Research. Systematic investigations of the low temperature region (23K-300K) reveal a surprisingly rich physics of mesoatomic and mesomolecular processes, unparalleled in other systems of isotopic hydrogen mixtures<sup>10</sup>: A dramatic density dependence of the reaction rates is found. The rich structure in the time spectra of the fusion neutrons observed at low gas density yields first evidence for new effects, most likely strong contributions from reactions of hot muonic atoms. The important question of muon losses due to He sticking is investigated by different methods and over a wide range of tritium concentrations.

## II. MUON CATALYZED FUSION CYCLE

The physics taking place when a muon comes to rest in DT mixtures has been covered in a number of theoretical papers<sup>11-13</sup>, see also M. Leon at this workshop. Thus we limit the following discussion to the simplified scheme of Fig. 1.

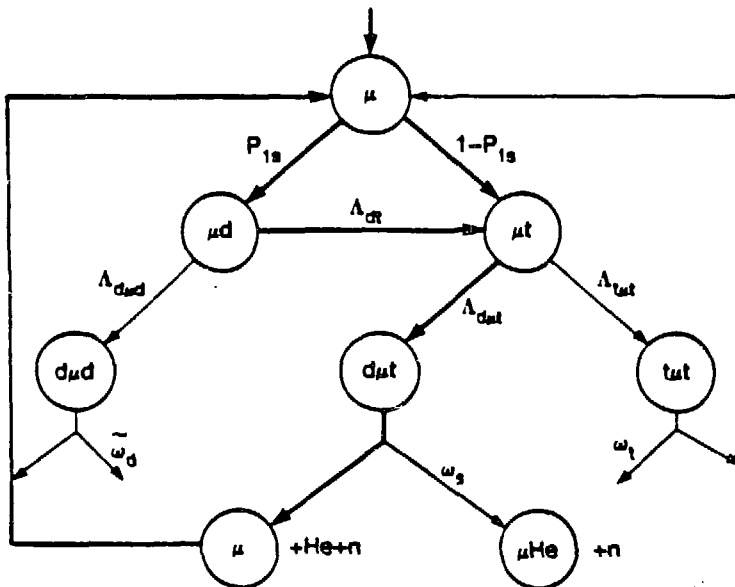


Fig. 1 Muon catalyzed fusion cycle in DT mixture

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After the initial muon capture into high atomic orbits, fast muon transfer from excited states  $\mu d^*$  to  $\mu t^*$  is expected to compete with cascade transitions to the ground state. This effect reduces the initial population  $P_{1s}$  of  $\mu d$  atoms in their ground state to

$$P_{1s} = c_d q_{1s} \quad (2)$$

where  $q_{1s} \leq 1$  strongly depends on the density  $\phi$  and tritium concentration  $c_t$ <sup>14</sup>. From the  $\mu d$  ground state isotopic transfer takes place with an effective rate

$$\Lambda_{dt} = \phi c_t \lambda_{dt} \quad (3)$$

All effective rates  $\Lambda_x$  depend on the target density  $\phi$ . The rates  $\lambda_x$  are normalized to liquid hydrogen density  $\phi_0 = 4.25 \cdot 10^{22}$  atoms/cm<sup>3</sup>. Gas densities are given relative to  $\phi_0$ .

In collisions with nuclei of the D<sub>2</sub>, DT and T<sub>2</sub> target molecules muonic molecules d<sub>μd</sub>, d<sub>μt</sub> or t<sub>μt</sub> are formed. Due to the resonance character of d<sub>μt</sub> formation, the rate  $\Lambda_{d\mu t}$  consists of strongly different contributions for molecular formation on D<sub>2</sub> and DT molecules<sup>11,13,15</sup>

$$\Lambda_{d\mu t} = \phi [2c_{D_2} \lambda_{d\mu t}^{D_2} + c_{DT} \lambda_{d\mu t}^{DT}] \quad (4)$$

( $c_{D_2}$  and  $c_{DT}$  denote the concentrations of D<sub>2</sub> and DT molecules, respectively,  $c_{D_2} + c_{DT} + c_{T_2} = 0.5$ ). Due to the large transition rate between  $\mu t$  hyperfine states<sup>16</sup>,  $\Lambda_{d\mu t}$  is expected to represent mainly d<sub>μt</sub> formation from the  $\mu t$  singlet state.

The behaviour of the effective d<sub>μd</sub> formation rate  $\Lambda_{d\mu d}$ , on the other hand, is dominated by more complex hyperfine effects at low temperatures<sup>2</sup>. (For the sake of simplicity the two hyperfine states of  $\mu d$  were not included explicitly in Fig. 1). Because the molecular formation rate from the upper  $\mu d$  hyperfine state is resonant at liquid temperatures,  $\Lambda_{d\mu d}$  is approximately proportional to the population of this state<sup>17</sup>:

$$\Lambda_{d\mu d} = 2c_{D_2} \lambda_{d\mu d}^{3/2} \frac{2}{3} \frac{\Lambda_{dt}}{\Lambda_{dt} + c_d \lambda_d^{\mu d}} \quad (5)$$

which leads to a nontrivial  $c_t$  dependence of  $\Lambda_{d\mu d}$  (for rate definitions see<sup>2</sup>).

In the various muonic molecules nuclear fusion occurs rapidly<sup>18</sup>. After fusion the muon is free to start a new cycle, except when it gets captured by the <sup>4</sup>He (or <sup>3</sup>He) fusion products with sticking probabilities  $\tilde{\omega}_d$ ,  $\omega_s$  or  $\omega_t$  (see Fig. 1).

### III. EXPERIMENTS

The measurements were performed at the  $\mu E4$  beam of the Swiss Institute for Nuclear Research. The experimental set up shown in Fig. 2 is a development from the apparatus used in our previous experiments (see<sup>2,8</sup> for more details).

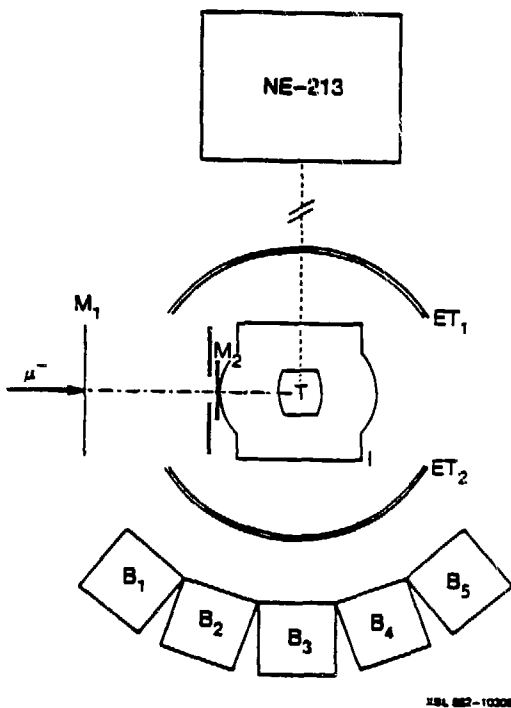


Fig. 2 Experimental setup:

Target (T)

Insulation Vacuum (I)

Muon Telescope ( $M_i$ )

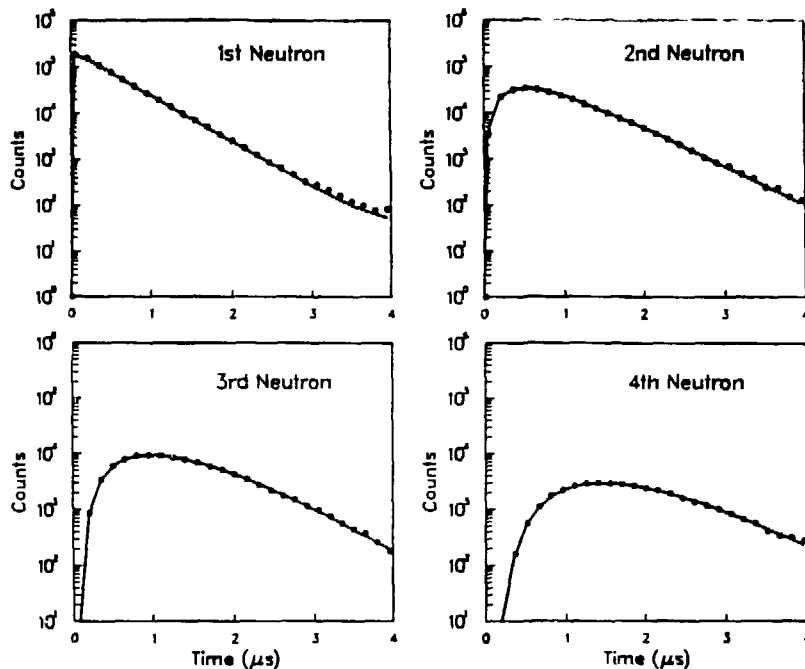
Neutron Detectors ( $B_i$  and NE213)

Electron Telescope ( $ET_i$ )

The DT liquid is contained in a cylindrical copper target cell ( $V=20\text{cm}^3$ ). Silver coating of the target walls and silver windows in the beam direction are used. (Muons stopping in silver disappear quickly by nuclear capture). The ratio of real stops to electronic stops varied during the measurements with liquid DT between 64% and 68%. For the experiments with gaseous DT mixtures larger targets ( $V=100-1000\text{cm}^3$ ) were used and effective stopping rates between 6% and 40% were achieved.

Time and energy spectra of neutrons (from dt and tt fusion) and electrons (from muon decay) were measured in 8 consecutive runs with high density liquid fillings ( $T = 23\text{K}$ , density 1.16-1.24) and in 32 runs with gaseous mixtures ( $T = 35\text{K} - 300\text{K}$ , densities from 0.5% to 8% of liquid hydrogen). The tritium concentrations were varied in a wide range from 2% to 96%.

As a significant modification to our previous set-up a set of 5 fast plastic counters (sizes 12" x 2" x 2" each) was installed to detect the large neutron multiplicities expected at high (liquid) target densities. Using fast routing circuits and pulse clipping techniques up to 4 subsequent neutron hits per detector were recorded. Due to the small efficiency ( $\epsilon_n = 4 \cdot 10^{-3}$ ) and dead time (50ns) per detector, pile up distortions of the time spectra were small and are well understood (Fig. 3). To determine the absolute neutron yields an experimentally calibrated liquid scintillation counter (5" x 4", NE213) with neutron-gamma pulse shape discrimination (PSD) was operated in addition. A sufficient distance (56cm) to the target was chosen to keep the occurrence of double neutron hits within the PSD integration time well below 10%. Systematic effects were also checked by doubling this distance at one run condition.



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Fig. 3 Time spectra of fusion neutrons observed subsequently in one of the plastic detectors at  $c_t = 0.36$ , liquid target. Solid curve demonstrates agreement with analytical expressions derived for these distributions including small effects of deadtime and accidentals.

The trigger for accepting events was a muon stop signal (with pile up rejection of beam particles entering the target area within  $9\mu s$  before and after) accompanied by at least one neutron or electron telescope signal within  $8\mu s$ . This simple and nonrestrictive trigger turned out to be valuable, since coincidence conditions between neutrons and electrons can lead to significant distortions of the neutron time spectra<sup>19</sup>. The full information about unconstrained neutron and electron time spectra allowed a careful off-line study of systematic effects and gave sufficient redundancy in the analysis.

One particular problem of DT fusion experiments is the handling of large quantities of tritium. For this purpose a closed loop high vacuum and gas filling system was constructed using exclusively metallic components and palladium filters for gas purification<sup>20</sup>. Different filling procedures enabled us to investigate equilibrated as well as non equilibrated molecular mixtures at liquid temperatures and to directly observe the effect of molecular concentrations on the  $d_{DT}$  formation rates (see Eq. 4). A mass spectrometer was connected to the target cell for an in situ determination of the molecular compositions.

## IV. ANALYSIS AND RESULTS

### A. Basic Kinetic Parameters

#### Steady State Behavior

Due to the strong coupling between the different states  $\mu d, \mu t \dots$  in the MCF cycle shown in Fig.1, a steady state can be attained during the muon lifetime. Then the relative probability of the muon being in one of these states is constant and all states disappear with the same loss rate  $\lambda_n$ , which is also the neutron disappearance rate. Because the effective rates  $\Lambda_x$  in Fig. 1 strongly depend on density, the time scale for reaching the steady state will range from a few ns in liquid DT to some 100ns in gas of density 1%.

In the steady state the time distribution of observed DT fusion neutrons is simply<sup>7</sup>

$$\frac{dN}{dt} = \epsilon_n \lambda_c e^{-\lambda_n t} \quad (6)$$

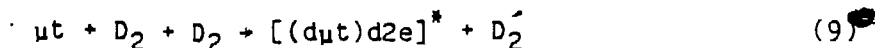
$$\lambda_n = \lambda_0 + W \lambda_c \quad (7)$$

where  $\lambda_c$  is the DT cycle rate, W the muon loss per cycle,  $\lambda_0 = 0.455 \mu s^{-1}$  the muon decay constant and  $\epsilon_n$  the neutron detection efficiency for 14 MeV fusion neutrons.

The inverse of the cycle rate  $\lambda_c$  characterizes the average time a muon needs to pass through the fusion cycle leading to DT fusion. It can be expressed in terms of the effective rates (Fig. 1)

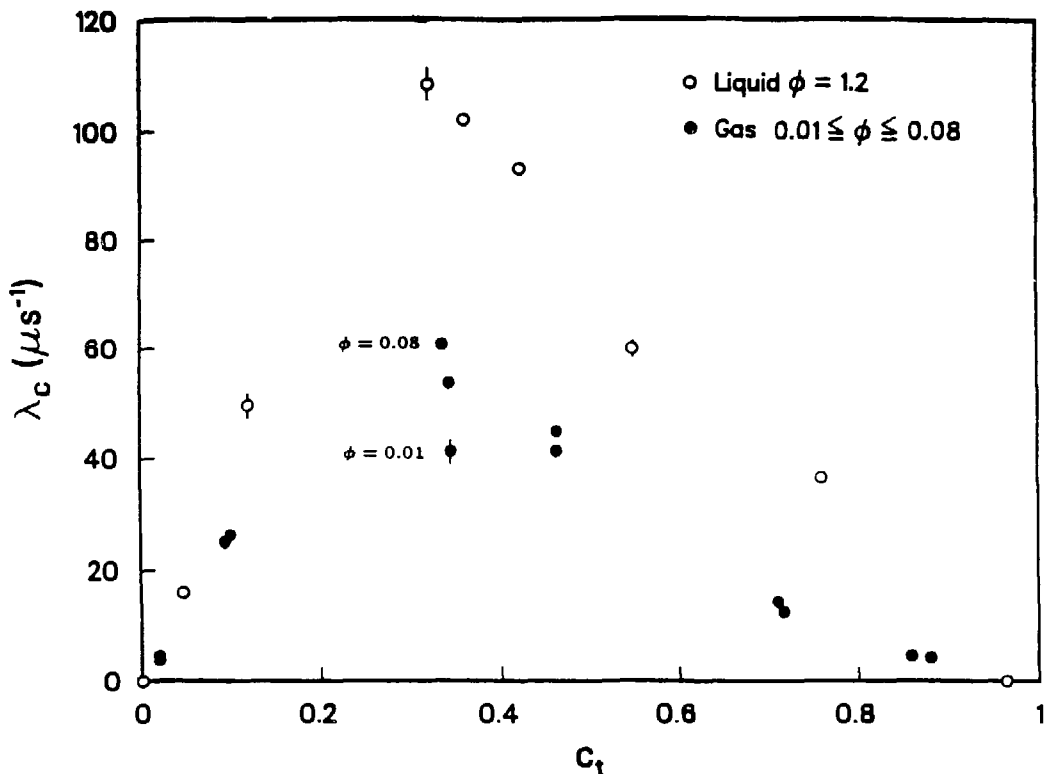
$$\frac{1}{\lambda_c} = \frac{P_{1s}}{\Lambda_{dt} + (1-P_{1s})\Lambda_{d\mu d}} + \frac{1}{\Lambda_{d\mu t}} \quad (8)$$

Figure 4 presents the cycle rates normalized to liquid hydrogen density observed in our experiments at low temperatures. Since these data were taken at nearly the same temperature (gas T = 35K, liquid T = 23K) temperature effects in the  $d\mu t$  formation rates should be insignificant. Thus the large difference between liquid and gas data seen in Fig. 4 is clear evidence for a surprising density dependence of the cycle rates which enhances the rates by a factor of ~2 at liquid conditions. Recent theoretical explanations<sup>21</sup> consider the effect of three body collisions in resonant  $d\mu t$  formation. If a small resonance defect exists for two body collisions, triple collisions like



with energy transfer to the spectator molecule allow large resonant rates. These conditions seem to be fulfilled for the dominating  $d\mu t$  formation rate from the singlet  $\mu t$  state, where the strongest resonances for two body reactions are below threshold<sup>22</sup>.





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Fig. 4 Cycle rates  $\lambda_C$  (normalized to LH<sub>2</sub> density  $\phi_0$ ) as function of  $c_t$  showing pronounced density effects.

A detailed analysis of the cycle rates  $\lambda_C$  obtained in liquid DT mixtures were performed in terms of basic rates. The observed cycle rates were fitted to Eq.(8) in the tritium range  $0.04 \leq c_t \leq 0.80$  using the measured atomic and molecular concentrations for the different data points. The results are presented in Table I together with preliminary values from the experiment at LAMPF<sup>23</sup>.

TABLE I	$q_{1s}$ Behavior	$\lambda_{dt}$ [ $\mu s^{-1}$ ]	$\lambda_{dt}^{D_2}$ [ $\mu s^{-1}$ ]	$\lambda_{dt}^{DT}$ [ $\mu s^{-1}$ ]
Theory <sup>13,14</sup>	drastic $c_t$ dependence drastic $\phi$	200		
Bystritsky, et al <sup>6</sup>	(not sensitive)	$290 \pm 40$	>100	(not sensitive)
Jones et al <sup>23</sup> preliminary T < 130K	Weak $c_t$ dependence no $\phi$	$284 \pm 40$ (T=23K)	$656 \pm 58$	$26 \pm 6$
This experiment preliminary T = 23K	strong $c_t$ dependence $\phi$	~ 250	$300 \pm 50$	$10 \pm 30$

A key problem for the analysis is the correct choice of  $q_{1s}$  (Eq.2), describing the fast muon transfer, which is expected to be a function of  $\phi$ , and  $c_t$ <sup>13,14</sup>. It is difficult to disentangle  $q_{1s}$  from the ground state muon transfer rate  $\lambda_{dt}$ , because at low  $c_t \leq 0.1$

$$\lambda_c = \frac{c_t \lambda_{dt}}{c_d q_{1s}} \quad (10)$$

The value of  $\lambda_{dt}$  adopted in table I results from preliminary analysis of our gas data at low  $\phi, c_t$  and our recent experiment with extremely low  $c_t \sim 4 \cdot 10^{-4}$  liquid targets, where all models predict  $q_{1s} \sim 1$ .

Also, one should note the large discrepancy concerning the rate  $\lambda_{dut}$ <sup>D2</sup>, which is a direct consequence of the different  $c_t$  dependence of cycle rates observed in the LAMPF data and this experiment.

### Transient Behavior

A more direct approach to disentangle the complex kinetics of muon induced processes is pursued in low density gas experiments. Because of the density dependence of effective rates due to collisions, fast rates (up to some  $1000 \mu s^{-1}$  at liquid conditions) become directly observable at low densities<sup>8</sup>. Figure 5 presents examples for the rich structure of neutron time distributions observed at  $\phi = 1\%$ . Two distinct components are seen. The intensity of the fast component is extremely high, corresponding to  $d_{ut}$  formation rates approaching  $1000 \mu s^{-1}$ . Our original interpretation of these data in terms of a hyperfine model resulted in significant disagreement with theoretical calculations as discussed already<sup>8</sup>. Only recently a new explanation of our data has been proposed<sup>24,25</sup>: The intense components correspond to molecular formation by hot, non thermalized  $\mu t$  atoms, the transient times represent the thermalization times at various tritium concentrations. Thus, these transients provide first evidence for largely enhanced molecular formation rates in an energy range corresponding to a few 1000K, which significantly exceeds the energies reached in hot target experiments so far.

### B. Sticking

Integration of the time distribution of fusion reactions Eq.(6) yields the average number of fusions  $Y$

$$Y^{-1} = \frac{\lambda_0}{\lambda_c} + W \quad (11)$$

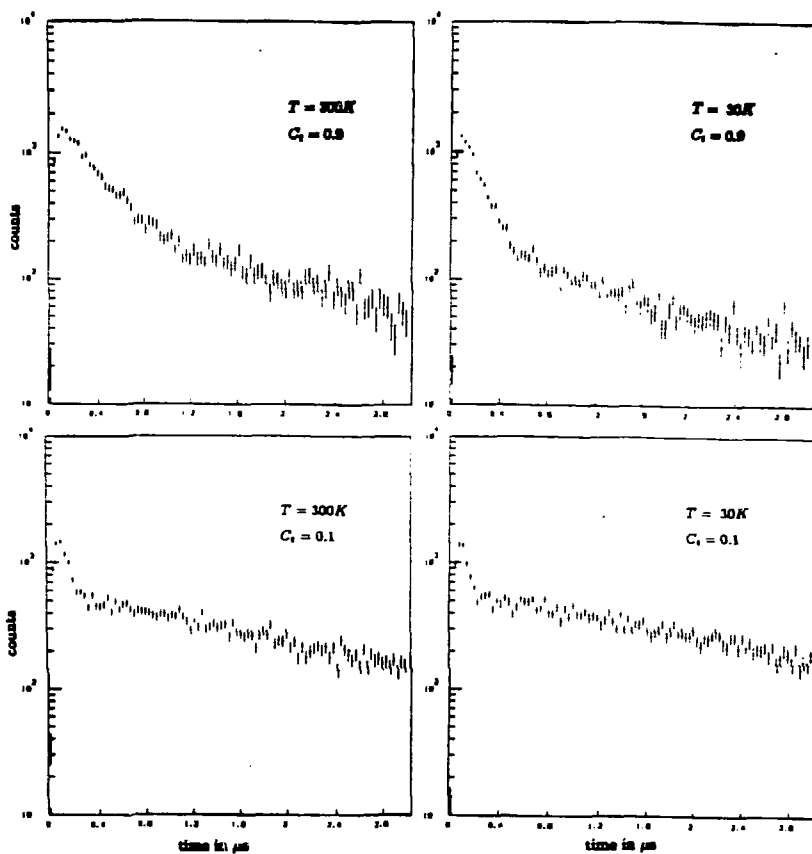


Fig. 5 Examples for time spectra of fusion neutrons at 1% gas density. The dependence of this time structure on  $c_t$ ,  $T$  and  $\phi$  gives direct information about the kinetic parameters.

The muon losses per cycle  $W$  can be further decomposed into<sup>26</sup>

$$W = \omega_S + \frac{\lambda_f^{tt} \Lambda_{tut} \omega_t}{(\lambda_f^{tt} + \Lambda_{tut}) \Lambda_{dut}} + \frac{P_{1s} \Lambda_{dud} \tilde{\omega}_d}{\Lambda_{dt} + (1 - P_{1s}) \Lambda_{dud}} + \dots \quad (12)$$

(for rate definitions see Fig.1.  $\lambda_f^{tt}$  is the fusion rate of  $t_{ut}$  molecules<sup>27</sup>)  
 The loss terms into competing fusion channels become less important with increasing  $\lambda_c$ . Thus, with sufficiently high cycle rates ( $d_{ut}$  formation rates of hot atoms observed are already ~ 2000 times faster than  $\lambda_0$ ) the intrinsic sticking of the  $d_{ut}$  fusion  $\omega_S$  ultimately limits the fusion yield catalyzed by a single muon to  $Y \leq 1/\omega_S$ .

### Neutron Method

Figure 6 shows the raw sticking values  $W$  obtained from the disappearance rates  $\lambda_n$  of fusion neutrons at liquid target conditions. Because preliminary LAMPF data for  $c_t \leq 0.3$  have indicated a surprising dependence of  $\omega_S$  on the tritium concentration<sup>28</sup>, the sticking probability  $\omega_S$  was evaluated over the whole range of tritium concentrations observed in our experiment. Once a set of kinetic rates is determined from the fit of the cycle rates, the contributions of the various loss channels can be subtracted from  $W$  to give  $\omega_S$

according to Eq. 12. The most important corrections are calculated from experimental results ( $t_{ut}$  rates from our data at  $c_t = 0.963$ ,  $\omega_d$  from<sup>29</sup>,  $\Lambda_{dud}$  including hyperfine effects from<sup>2,30</sup>,  $p_{ud}$  rates from<sup>31</sup>), only for  $p_{ut}$  molecular formation theoretical values<sup>32</sup> are used. The values of  $\omega_s$  resulting from this preliminary analysis show no significant  $c_t$  dependence<sup>33</sup> with

$$\omega_s = (0.50 \pm .08)\% \quad (13)$$

somewhat lower than most recent theoretical calculations<sup>34-37</sup>.

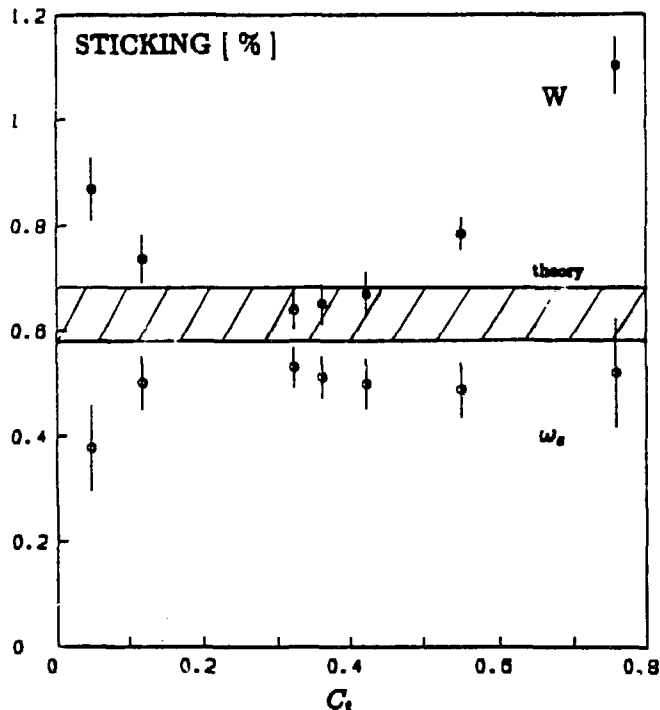


Fig. 6 Raw sticking  $W$  and sticking  $\omega_s$  (after subtraction of other loss channels). Dashed line shows current theoretical predictions<sup>34,36</sup>.

### X Ray Method

Additional detailed information about the sticking process can be obtained by observation of X-rays from excited  $\mu\text{He}$  states following fusion. The overall sticking probability  $\omega_s$  consists of

$$\omega_s = \omega_s^0 (1-R) \quad (14)$$

where  $\omega_s^0$  characterizes the initial sticking and  $R = 0.24-0.32$  is the reactivation efficiency of the muon during the slowing down of the recoiling  $\mu\text{He}$  ion<sup>31,37,38</sup>. Both these important steps, the initial population of  $\mu\text{He}$  levels<sup>34,35</sup> and the understanding of collisional processes<sup>36,37</sup>, are systematically studied by observation of X-ray intensities. Our results for  $p_{ud}$  and  $d_{ud}$  have been published<sup>39</sup>. An experiment in liquid DT ( $c_t = 4.8 \cdot 10^{-4}$ ), which is extremely difficult due to the high tritium activity, has been performed successfully. First, very preliminary results indicate a  $2p \rightarrow 1s$  yield of  $\sim 2 \cdot 10^{-3}$  per fusion, smaller than the recent theoretical value of  $3.6 \cdot 10^{-3}$ <sup>37</sup>.

## V. CONCLUSION

A systematic study of muon catalyzed DT fusion at low temperature has produced a consistent set of data showing high cycle rates (see Fig. 4) and very small DT sticking values ( $w_s \approx 0.5\%$ , see Fig. 5). Our maximum observed neutron yield per muon just exceeds 100 and could approach 200 if conditions with even large cycle rates can be achieved. Indeed, our gas data indicate a sharp rise of fusion rates in the case of hot, non thermalized  $\mu t$  atoms (see Fig. 5). Thus, at high temperatures, conditions for extremely high fusion yields may be found.

Our observations can be described in terms of the kinetic model displayed in Fig. 1 and outlined in Section II and IV. A non trivial density effect of the cycle rate between gas and liquid data is well established over the whole range of investigated tritium concentrations. It can be understood qualitatively by two different mechanisms: at low  $c_t$  by isotopic exchange during the muonic cascade<sup>14</sup> and at  $c_t \geq 0.15$  by resonant  $d\mu t$  formation in triple collision<sup>21</sup>.

A comparison with the experiment at LAMPF<sup>23</sup> shows agreement in the magnitude of  $\lambda_c$  and  $w_s$  observed, but discrepancies up to a factor of 2 in the resonant  $d\mu t$  formation rate and qualitatively different behavior of  $q_{1s}$ , i.e. isotopic transfer from excited states (see Table I). Our preliminary analysis indicates no significant  $c_t$  dependence of sticking  $w_s$ , see Fig. 6.

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